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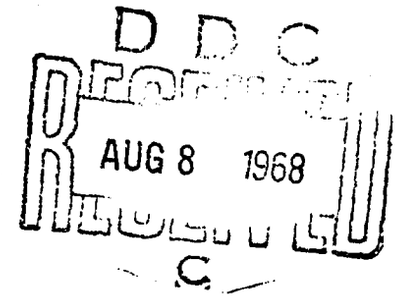


Advances in Aerobiological Aerosol Research. per Mrs. Anderson O.R.C. 15 Aug 68

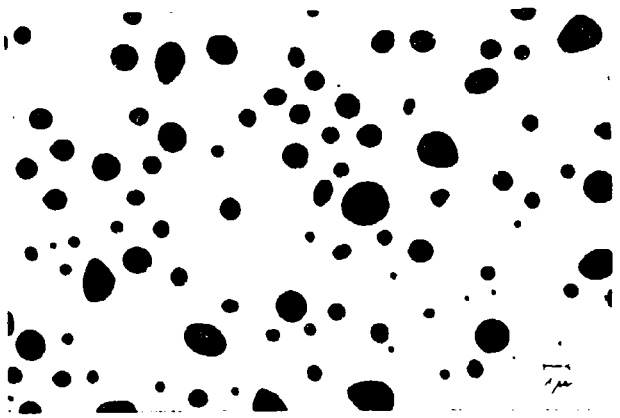
DISCUSSION

Fortschritte der biologischen Aerosol-Forschung-Jahren 1957-1961, pp 297-303

Pfefferkorn, Munster:

These condensations of droplets on small particles can also be observed in the electron microscope under certain conditions. To do this, we get the electron microscope object carrier (foil, net edges, or oxide needles with magnesium oxide which is precipitated in the form of small cubes). We irradiate these prepared object carriers with light, ultraviolet or x-rays in air which contains corresponding hydrocarbons with oxidizing gases and water vapor; if we do this, we find that droplets are formed around the little cubes and these droplets increase in size as we continue the irradiation. The illustration shows droplets of this kind on a Formvar foil with MgO-crystals which were irradiated for 5 hours in benzol steam with soft x-rays. These little droplets -- contrary to the assumption of Dr. Cauer -- cannot contain unbound water for they are stable in the high vacuum of the electron microscope and under electron bombardment. (Lecture at the Conference of the German Association of Electron Microscopy, 1957, Darmstadt, Phys. Verhandlungen (Physical Transactions), 8, 239, 1957.)

I would just like to make one more remark on the vaporization speed of the very small droplets. The smaller the droplets are, the faster they are vaporized because the vapor pressure increases. We cannot confirm this theoretically-founded assertion in our practical experiments. Using diocthylphthalate and various oils, we found that small, ball-shaped droplets, speared on oxide needles, are much more stable in the high vacuum than when they were precipitated on a foil in the shape of hemispheres. For instance, droplets cannot be studied on foil in the electron microscope, whereas they can be shown as little round balls on the oxide needles. The steam pressure increase, which we get when the radius of the ball is smaller, thus does not in each and every case lead to faster vaporization.



MgO-crystals on Formvar foil, irradiated for 5 hours with x-rays in benzol steam. Magnification: 5000:1.

Goetz, Pasadena:

Let me just say that I am fully aware of what my colleague Pfefferkorn just told us. I was talking about more volatile combinations, which form photochemically developing condensates, which in turn can no longer be caught in the electron microscope. Here we can refer, for instance, to the studies by Busch and Robertson in Los Angeles; these two researchers have tried to catch the smog, such as it actually develops, in the form of precipitation; they were actually trying to interpret the smog with the help of the electron microscope. They found that this is impossible. In case of less volatile substances or in case of heavy hydrocarbons, for instance, in the oxidizing terpenes, we can accurately observe the formation of the condensate. But we are afraid that it is precisely the lighter hydrocarbons which play a role in this smog; here we would have the pentenes, the hexenes, and the heptenes. It appears that the molecular weights of the oxydants, which develop out of this, are not large enough to be prevented from becoming vaporized under the conditions prevailing in the electron microscope.

Cauer, Bochum:

Professor Pfefferkorn is entirely correct. The condensation method definitely does not refer only to substances which have a hygroscopic character. And the hygroscopic character is after all

always determined by the various laws of approximate forces. Where these forces are not present, such as in oil, for instance, this is of course entirely out of the question.

von Eichborn, Neu-Gilching:

Earlier, A. Goetz, H. Cauer, and I (Kolloid-Z. (Colloid Journal), 164, 41, 1960; Z. Aerosol-Forsch. (Aerosol Research Journal), 3, 279-374, 1954 as well as in analogy to similar problems in suspensions and liquids) assumed that, in the association of molecules in the vicinity of the surface of aerosol particles, we can also expect to find a loose aggregation having a relative density which is far smaller than the density of the condensed liquid. The comment on tobacco smoke by Professor Schmidt -- to the effect that there is a continual transition between the gas phase and the particle phase -- pretty much checks with my observations on the sedimentation velocity of tobacco fog and its increase under the effect of light, though this was also found, for instance, in the case of diesel oil fog.

Professor Goetz was quite correct in using the term "deposit" because the word "adsorption" generally conveys the idea of a surface condensation along the layers which would then be equivalent to the density of the liquid. We will have to devise other methods if we want to use the aerosol centrifuge mass spectrometer in breaking down the combination of equivalent diameter and relative density into these two components -- that is, the combination which can be determined only from the sedimentation in the centrifugal field according to Stokes-Cunningham. It would seem that we can demonstrate, in the electron microscope, only cases of genuine "vapor concentration" which would be temperature-stable.

Goetz, Pasadena:

Mr. von Eichborn is entirely correct in saying that we could make measurements only at one temperature figure. But we can make measurements also at two different temperatures. And I would like to say also that we can make measurements isothermically at exactly the same temperature that we find in the environment; we can do this also under conditions where our temperature is 10° higher or 10° lower than this. The stability of the condensate, of the associated cloud, or call it what you will, is clearly expressed by the thermal sensitivity. If everything is completely stable, then 10° would hardly make any difference. The shrinkage of the diameter or the increase in the diameter is constant and depends on the temperature. But where we are dealing with weak associations, the change in the diameter turns out to be dependent on the temperature. In this manner we hope that we will be able to arrive at a correct conclusion as to the density.

Flach, Davos:

The intensification of research toward submicron aerosols and their photochemical and synergical effects call for a detailed investigation of the micro-and macrometeorological (turbulence, exchange of the variance of the irradiation intensities of sun and sky, and so on) -- of course, in those instances where we are investigating aerosols in the open air. In addition we would have to take into consideration the climatic conditions in our aerosol studies. Furthermore we must devote special attention to the manifold changes in water vapor aerosols by looking at the fluctuations in water vapor pressure and the relative humidity; this is indicated particularly on the basis of our own investigations.

Neuwirth, Freiburg:

The Freiburg (Breisgau) weather bureau has a biometeorological branch which is studying the problem of air pollution. However, this outfit is making measurements only in support of weather forecasts for our health resorts. For this purpose, an entire network of measurement facilities was set up, extending from the Allgaeu section via the Black Forest all the way to the Tauber River. The measuring instruments here are aluminum foil according to Diem and glass filter pots according to Teichert. In two places, they have an avigraph according to Effenberger. The comparatively pure air of the Black Forest reveals that there is a connection between the weather situation, the layers in the atmosphere, and the movement of weather fronts. The average dust load in out-of-the-way health resorts is still small and in most cases does not exceed 1 g/sq m/30 days. But many other health resorts have places where the dust is much more voluminous. These places are now being recorded and the health resort managements are being told how important it is to keep the air clean in the interest of the future development of the health resort itself; this is very important with regard to bioclimatic considerations.

Bisa, Grafschaft (County):

There are two questions which I would like to ask Dr. Schwarz:

Here is my first question: The determination of the micro- and the macro-values essentially is based on investigation results which in my opinion are somewhat incomplete. Usually, we take as our test object a plant or various species of plants which we assume are particularly sensitive to a certain aerosol. But we are of the opinion that it is not the plant or any kind of test model that constitutes a decisive criterion for the biological effectiveness of such aerosols and also for the determination of their micro- or macro-boundaries; this applies particularly to the radioactive aerosols; we say that we must also have test objects which are really strong and resistant in the biological

sphere so that we would get micro- and macro-limits that are meaningful as far as man is concerned.

Here is my second question: We know, on the basis of the synergisms of certain aerosols that, for instance, an aerosol or component A is completely inert and that the same is true of an aerosol component B. Now, if A and B get together, we get an aerosol C as a result of physical or chemical changes; this aerosol C may be highly toxic. We therefore must ask ourselves whether we have a concommitant causal relationship here, that is, whether industrial plant A, B, C, or D or other factories in the area are all contributing to the pollution of the air. We must have a way of finding the principal sources for these toxic synergisms in order to attack them first.

Wentzel, Bochum:

I must disagree with Dr. Schwarz' statement to the effect that "we can keep the neighborhood clean" if we stay within the micro-values. The micro-values, as we know, are measured as half-hour average values (and therefore represent integrals); this means that they will contain higher and at times toxic peaks. They do not correspond to the genuine toxicity or tolerance limits. This means, that we may still get damage even if we do adhere to our micro-values. Furthermore, the concept of "permissible" should be explained further. It would lead to misunderstandings if this term were used in a very general sense. Something that might possibly be declared permissible from the technical viewpoint need not necessarily be also permissible from the legal or hygienic viewpoint. The same also applies to the term "acceptable." It is not up to the VDI to say what is "acceptable" as regards health and so on.

Schwarz, Essen:

Gentlemen, I believe you can tell from my lecture how heterogeneous this entire field is; I think you will see that even a lecture lasting several hours would not have been long enough to cover this field completely. I therefore had to keep this brief and I could not go into detail in my discussion of the first question as to the process involved in the determination of the micro-values for SO_2. To this end, we have farmed out a series of research projects; an experimental station was set up in Siegerland (Biersdorf); we set up experimental stations in an open-air facility at varying distances from an emmitter -- an ore roasting installation which essentially emits SO_2. Every experimental station was provided with plants which represented a cross section of particularly important crops (grain, vegetables, fruit trees, deciduous and evergreen trees). We made this selection because it turned out that the resistance series which we have available in our literature unfortunately could not be used. The experiments were continued over a number of years. We were able to observe the damage and we recorded the SO_2 concentrations as we went along and all material was very carefully evaluated.

This project was handled by a research team consisting of Professor Dr. Egle, Frankfurt University, as scientific director and Dr. Stratmann, Essen, as well as Dr. Zahn, Frankfurt-Hoechst, and of course Dr. Guderian, who was in charge in field work. Parallel to this effort, Dr. Zahn made open-air experiments in Hatterscheim near Hoechst; the Hoechst Dyestuff Plant financed this project; here, the same species of plants were subjected to short-peak gassing and were then again placed in a normal environment. Third, Dr. Stratmann conducted an experimental program in controlled climate chambers in Essen, where the plants were exposed to certain types of gasses under well defined conditions. On the basis of these three components we then determined the damage limits for gassing with SO_2. We have a series of publications on this (see also the bibliographic references (9) to (13) in the original essay). We have numerical material amounting to several hundred thousand data which are now being further evaluated because we of course obtained a whole series of supplementary results, especially in connection with resistance.

We are also of the opinion that such problems must be tackled very carefully and that we cannot really define a boundary value until we have it carefully checked by all people involved, that is doctors, veterinarians, botanists, forestry scientists, and engineers. In addition to the researchers mentioned above, we are particularly indebted to Professor Neumann who gave us valuable advice on the human medicine aspects of this problem.

Similar experiments are now going to be conducted with fluorine because this problem is likewise very complicated and difficult. We are planning a series of research projects for fluorine; this is once again going to be very expensive.

Now, let us take up the second problem, which concerns synergisms. I emphasized that we started here with the simple problems. The synergistic effects of several pollution components will be taken up by our committee on "metropolitan air," which has started its preliminary preparations along these lines. We are of course quite aware that we will have combined effects in many cases. But we also have a series of towns -- particularly in the Ruhr region -- where we essentially need to expect only SO_2. In addition, we of course have many places where we simultaneously encounter, for instance, nitric oxide and sulfur dioxide. This combination is being tested by Dr. Stratmann in a separate series of experiments, again in air-conditioned rooms. Other laboratories are making very detailed tests with soot, that is to say, with soot not containing sulfur dioxide and with soot containing deposited sulfur dioxide. We think that it will take quite some time before we can track down these very complicated interrelationships.

Neumann, Wuertburg:

The factors which must be considered from the medical viewpoint have of course been given full consideration in the establishment of the micro-values for SO_2. In order to answer questions concerning the odor and irritation threshold SO_2 the Institute of Pharmacology and Toxicology of the University of Wuertburg has conducted a special series of experiments. All of the important meetings of the committee on pure air were attended by doctors as well as botanists, forestry experts, analysts, engineers, and veterinarians. One thing that is particularly important here is the British report which was mentioned by Dr. Schwarz and which says that certain illnesses are aggravated whenever an increase in the daily average SO_2 to 0.4 ppm coincides with greatly increased dust content in the air. According to the current state of science, we can say that the sustained and short-term micro-values for SO_2 do not constitute a danger to man; these, by the way, are the values Dr. Schwarz gave for West Germany.

Bergerhoff, Bochum:

Dr. Schwarz explained the term micro-value -- in contrast to what we generally and internationally mean by it -- by saying that the term actually designates the still acceptable harmful substance limit value in the air. On the basis of this explanation, one could hardly say that this involves an effort to find the normal micro-values. Looking at it from the angle of the actual effect, this interpretation would imply going into the damage region of substances that are foreign to air. The acceptability factor, according to Article 906, BGB (Federal Legal Gazette), by the way, comes under an entirely different heading.

Some of the definitions so far suggested for our micro-value criteria are too inexact to give us effect boundary values. Accordingly, we may often have a possibility of intermediate deviations from the actually observed values and this of course would have detrimental effects; here we get into the entire issue of responsibility. I think that we ought to be highly concerned over the fact that the air purification commission of the VDI is still suggesting so-called permissible immission values over a period of half an hour, without there being any need for such measurements. As we know, the immission varies considerably at the same place within this span of time and the concentration of substances foreign to air varies also; at the peak we may have a situation where the immission value, which otherwise might still look harmless and acceptable, would actually exceed the tolerable value many times.

Quite often, the concentration threshold, which is supposed to indicate the micro-value, is also expressed with the term tolerance

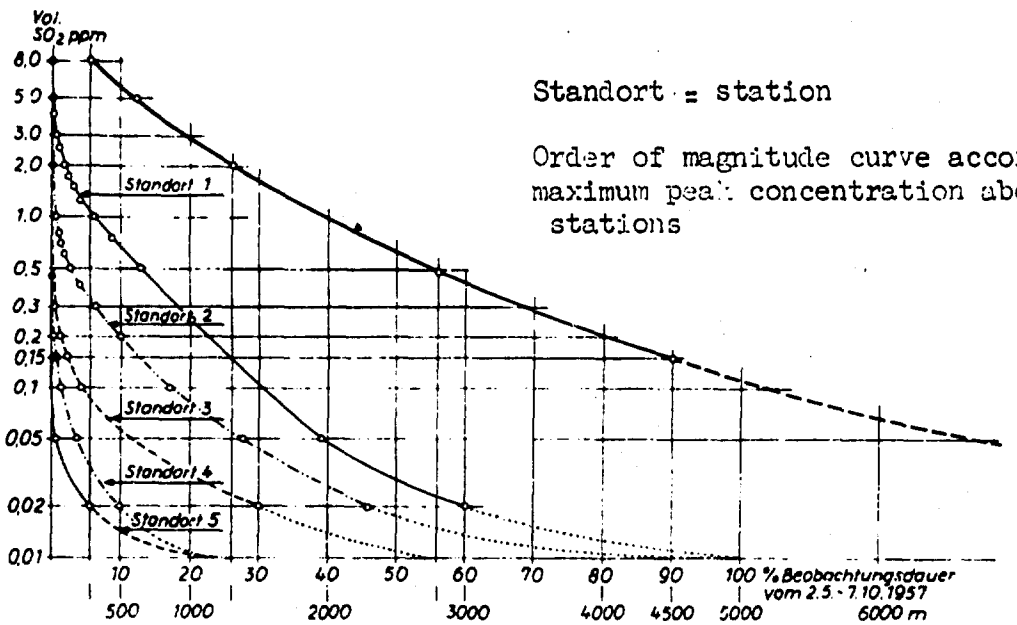
limit. Both terms are supposed to express the dividing line between dangerous and harmless substances in the air. Of the micro-values we expect that they will indicate the critical boundary at which we can anticipate harmful biological effects. We must absolutely stay within this boundary so that we may exclude certain negative effects from our considerations. We can exclude these values and these effects only when we do not at any time exceed the critical boundary in the area where we do have immission.

Dr. Schwarz also mentioned the "Biersdorf experiments" which were part of the research project on "SO_2 Effects on Vegetation." Here the main job was to use pot cultures in open-air conditions in order to provide material which would help us determine the concentration threshold at which the biologically harmful effects of SO_2 in the air set in. He emphasized only the research phase from 1958-1960 where Dr. Stratmann and associates (Air Purification Research Institute, Incorporated, Essen) did all the work. However, he did not discuss the prior research in 1957. This was the phase during which the first actual observations were made. The meteorological and gas-analysis portion of this project was handled by Essen Technical Surveillance Committee, Incorporated (Dr Schwarz and associates), while the biological portion was handled by the Soil Improvement Institute in Bochum (Dr. Bergerhoff and associates). We must take a close look at the findings of 1957 because the SO_2 measurements, using Woesthoff equipment in several places in the terrain, during this period were made at greater distances from major SO_2 emitters, that is, the ore roasting installation in Biersdorf, than during the period from 1958 to 1960. The continual SO_2 control in air near the ground from 1958 to 1960 was based only on five points in the terrain which were about 325, 600, 700, 1300, and 1900 m away from the emitter, whereas in 1957 these measurements were taken in five places over a period of 5 months where the distances from the emitter, moving toward the east, were about 275, 600, 1300, 2800, and 4500 m.

The evaluation of data, which we have for the measurement points of 1957 with regard to the SO_2 content of the air, as well as my supplementary observations on damage due to smoke gas effect on open-air flora, outside the area of the experimental stations, quite clearly indicates that the biological-toxic boundary value for SO_2 should be found approximately in the concentration range having an order of magnitude of about 0.1 Vol ppm. This figure can be confirmed by the fact that we were able to find fir trees considerably damaged by smoke about 4.5 km from the roasting kiln of the Fuesseberg mine and Biersdorf. There was definitely a connection between the macroscopically visible damage on the trees from such as thin needle coverage and loss of needle growth, as well as the decrease in the SO_2 effect traces along the air currents flowing over the Biersdorf

or roasting facility and the windward slopes leading to the place where the trees stood. As far as location is concerned here we can say that the SO_2 peak concentration in this part of the terrain will probably have an even lower value than the value we were able to find at the station which was about 4.5 km from the Biersdorf roasting kiln; here the air near the ground had for a long time been checked for its SO_2 content. At this place, we measured a maximum peak concentration of 0.1 Vol ppm during only several minutes duration from the start of May to the beginning of October 1957.

We get the following if we plot the SO_2 curve for 1957 in such a manner that we get a series of values arranged according to size and time.



Standort = station

Order of magnitude curve according to maximum peak concentration above stations

100% observation time from 2 May - 7 Oct 1957

Graph of SO_2 recorded at plant observation station along the main wind direction behind the ore roasting kilns in Biersdorf (Wester Forest) based on data collected (a) from 2 May - 7 October 1957, (b) 1957-1959 (as regards station 1 and station 2, we considered the data from the years 1958-1959).

1. At each measuring point, the incidence of SO_2 on the whole followed an exponential law.

2. The peak concentration obtained at the various control points, as the distance of the measuring points from smaller emmission sources increased, assumed smaller values and the line in general approached a terminal value asymptotically.

After 1957, it appears that we had similar SO_2 incidence conditions in the distance leeward effect area of the Biersdorf roasting installation; we say that because we measured a smoke source distance of up to 1900 m, at SO_2 concentrations comparable to 1957, though we used the same control point of the immission area.

The phenomena we observed in forests which were 45 km and further from Biersdorf plus the fact that the diffuse smoke incidence from the direction of Biersdorf takes effect there but since the concentration peak according to past observation here, by the way, is 0.1 Vol ppm would seem to indicate that this figure should be the immission boundary value for SO_2.

Schwarz, Essen:

The problem which Dr. Bergerhoff brought up here was discussed in various committees of the "VDI Air Purification Commission." You will find these questions answered in VDI Memorandum 2108 entitled: "Maximum Immission Concentrations (Micro Values) Sulphur Dioxide" (see bibliographic reference 13 of the original essay) and in the bibliography given there.